

WHAT IS CLAIMED IS:

1. A method for crystallizing epsilon polymorph 2,4,6,8,10,12-hexanitro-2,4,6,8,10,12-hexaazatetracyclo[5.5.0.0<sup>5,9</sup>0<sup>3,11</sup>]-dodecane (CL-20), comprising:

preparing a substantially dry CL-20 solvent solution containing an amount of CL-20 dissolved in a CL-20 solvent;

providing a crystallizer containing a CL-20 non-solvent;

adding the substantially dry solvent solution to the crystallizer containing the CL-20 non-solvent to cause precipitation of epsilon polymorph CL-20 crystals by inverse precipitation technique; and

separating the precipitated epsilon polymorph CL-20 crystals from the non-solvent and the solvent.

2. A method according to claim 1, wherein said preparing of the substantially dry CL-20 solvent solution comprises substantially drying a wet CL-20 solvent solution containing the amount of CL-20 dissolved in the CL-20 solvent.

3. A method according to claim 2, wherein said substantial drying of the wet CL-20 solvent solution comprises azeotropic distillation to remove an azeotrope comprising water and the CL-20 solvent.

4. A method according to claim 1, wherein the substantially dry CL-20 solvent solution contains less than 1.5 weight percent water.

5. A method according to claim 1, wherein the CL-20 solvent comprises at least one member selected from the group consisting of ethyl acetate, methyl acetate, isopropyl acetate, butyl acetate, tetrahydrofuran, and methyl ethyl ketone.

6. A method according to claim 1, wherein the CL-20 solvent comprises ethyl acetate.

7. A method according to claim 1, wherein the solubility of CL-20 in the solvent is greater than 20 percent weight/volume (g/ml).

8. A method according to claim 1, wherein the CL-20 non-solvent is free of halogens.

9. A method according to claim 1, wherein the CL-20 non-solvent is free of chlorine.

10. A method according to claim 1, wherein the CL-20 non-solvent comprises at least one member selected from the group consisting of hexane, cycloheptane, heptane, octane, benzene, toluene, and xylene.

11. A method according to claim 1, wherein said separating of precipitated epsilon polymorph CL-20 crystals from the non-solvent and the solvent comprises filtration.

12. A method according to claim 1, wherein the precipitated epsilon polymorph CL-20 crystals comprise particles having maximum diameters of, on average, about 40  $\mu\text{m}$  to about 70  $\mu\text{m}$ .

13. A method according to claim 1, further comprising adding a co-non-solvent to the wet CL-20 solvent solution or the substantially dry solvent solution, the co-non-solvent comprising at least one member selected from the group consisting of naphthenic oil, paraffinic oil, benzyl formate, and poly(propylene glycol).

14. A method according to claim 13, wherein a weight ratio of co-non-solvent to non-solvent is in a range of from about 5:95 to about 20:80.

15. A method according to claim 1, further comprising preparing the CL-20 from 2,6,8,12-tetraacetyl-2,4,6,8,10,12-hexaazatetracyclo-[5.5.0.0<sup>5,9</sup>0<sup>3,11</sup>]-dodecane (TADA).

16. A method according to claim 1, further comprising, subsequent to said separating, washing the precipitated epsilon polymorph CL-20 crystals with at least one member selected from the group consisting of isopropanol and ethanol, and thereafter washing the precipitated epsilon polymorph CL-20 crystals with water.

17. A method for crystallizing epsilon polymorph 2,4,6,8,10,12-hexanitro-2,4,6,8,10,12-hexaazatetracyclo[5.5.0.0<sup>5,9</sup>0<sup>3,11</sup>]-dodecane (CL-20), comprising:

dissolving an amount of CL-20 into a solution containing a CL-20 solvent and water to form an aqueous phase and a wet CL-20 solvent phase, wherein the CL-20 is dissolved in the wet CL-20 solvent phase;

substantially drying the wet CL-20 solvent solution to thereby form a substantially dry solvent solution containing the CL-20;

adding a base to the CL-20 solvent phase to neutralize acidic species;

providing a crystallizer containing a CL-20 non-solvent;

adding the substantially dry solvent solution to the crystallizer containing the CL-20 non-solvent to cause precipitation of epsilon polymorph CL-20 crystals by inverse precipitation technique; and

separating the precipitated epsilon polymorph CL-20 crystals from the non-solvent and the solvent.

18. A method according to claim 17, wherein the base comprises at least one member selected from the group consisting of Na<sub>2</sub>CO<sub>3</sub>, K<sub>2</sub>CO<sub>3</sub>, NaHCO<sub>3</sub>, KHCO<sub>3</sub>, NaOH, and KOH.

19. A method according to claim 17, wherein said substantial drying of the wet CL-20 solvent solution comprises azeotropic distillation to remove an azeotrope comprising water and the CL-20 solvent.

20. A method according to claim 19, wherein the dry CL-20 solvent solution contains less than 1.5 weight percent water.

21. A method for crystallizing epsilon polymorph 2,4,6,8,10,12-hexanitro-2,4,6,8,10,12-hexaazatetracyclo[5.5.0.0<sup>5,9</sup>0<sup>3,11</sup>]-dodecane (CL-20), comprising:

preparing a substantially dry CL-20 solvent solution containing an amount of CL-20 dissolved in a solvent;

providing a crystallizer containing a CL-20 non-solvent and seed crystals of epsilon polymorph CL-20;

adding the substantially dry solvent solution to the crystallizer containing the CL-20 non-solvent and the seed crystals to cause precipitation of epsilon polymorph CL-20 crystals by inverse precipitation technique; and

separating the precipitated epsilon polymorph CL-20 crystals from the non-solvent and the solvent.

22. A method according to claim 21, wherein said preparing of the substantially dry CL-20 solvent solution comprises substantially drying a wet CL-20 solvent solution containing the amount of CL-20 dissolved in the CL-20 solvent.

23. A method according to claim 22, wherein said substantial drying of the wet CL-20 solvent solution comprises azeotropic distillation to remove an azeotrope comprising water and the CL-20 solvent.

24. A method according to claim 21, wherein the dry CL-20 solvent solution contains less than 1.5 weight percent water.

25. A method according to claim 21, wherein the CL-20 solvent comprises at least one member selected from the group consisting of ethyl acetate, methyl acetate, isopropyl acetate, butyl acetate, tetrahydrofuran, and methyl ethyl ketone.

26. A method according to claim 21, wherein the CL-20 solvent comprises ethyl acetate.

27. A method according to claim 21, wherein the solubility of CL-20 in the solvent is greater than 20 percent weight/volume (g/ml).

28. A method according to claim 21, wherein the CL-20 non-solvent is free of halogens.

29. A method according to claim 21, wherein the CL-20 non-solvent is free of chlorine.

30. A method according to claim 21, wherein the CL-20 non-solvent comprises at least one member selected from the group consisting of hexane, cycloheptane, heptane, octane, benzene, toluene, and xylene.

31. A method according to claim 21, wherein said separating of precipitated epsilon polymorph CL-20 crystals from the non-solvent and the solvent comprises filtration.

32. A method according to claim 21, wherein the precipitated epsilon polymorph CL-20 crystals comprise particles having maximum diameters of, on average, about 40  $\mu\text{m}$  to about 70  $\mu\text{m}$ .

33. A method according to claim 21, further comprising adding a co-non-solvent to the wet CL-20 solvent solution or the substantially dry solvent solution, the co-non-solvent comprising at least one member selected from the

group consisting of naphthenic oil, paraffinic oil, benzyl formate, and poly(propylene glycol).

34. A method according to claim 33, wherein a weight ratio of co-nonsolvent to non-solvent is in a range of from about 5:95 to about 20:80.

35. A method according to claim 21, further comprising preparing the CL-20 from 2,6,8,12-tetraacetyl-2,4,6,8,10,12-hexaazatetracyclo-[5.5.0.0<sup>5,9</sup>0<sup>3,11</sup>]-dodecane (TADA).

36. A method according to claim 21, further comprising, subsequent to said separating, washing the precipitated epsilon polymorph CL-20 crystals with at least one member selected from the group consisting of isopropanol and ethanol, and thereafter washing the precipitated epsilon polymorph CL-20 crystals with water.